Nonadiabatic dynamics and coherent control of nonequilibrium superconductors

Andreas Schnyder

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in collaboration with:

Holger Krull, Adolfo Avella, Dirk Manske Götz Uhrig, Alireza Akbari, Nikolaj Bittner, Ilya Eremin

Outline

1. Introduction

- Recent pump-probe experiments on NbN films
- How does a quantum system relax?

2. Nonadiabatic dynamics of superconductors

- Superconductor coupled to laser field and optical phonons
- Density matrix formalism

3. Results & Discussion

- Response of superconductor: Damped order parameter oscillations
- Generation of coherent phonons in non-equilibrium SC state: beating phenomena, resonant generation of coherent phonons
- Experimental signatures: Oscillations of pump-probe conductivity

4. Conclusions & Outlook

Motivation I: Recent pump-probe experiments



- THz-pump THz-probe spectroscopy:
- THz-pump pulse: Excite with intense femto-second pulse, induce dynamics
- THz-probe pulse: After delay time δt , measure with second, less intense pulse

> Optical conductivity / transmitivity as a function of ω and δt gives information about:

- Dynamics of SC condensate, order parameter oscillations
- Cooper pair recombination / recovery dynamics
- Coherent phonon oscillations

Motivation I: Recent pump-probe experiments

Matsunaga, Shimano, et al. PRL 109, 187002 (2012);PRL 111, 057002 (2013)

- THz-pump THz-probe spectroscopy on NbN films
- pump-pulse duration: 90 fs

non-adiabatic excitation of SC:

- $\tau_p \ll \tau_\Delta \sim h/(2|\Delta|)$
- Measure change in transmission of probe field $\,\delta E$

Observation:

- algebraically damped oscillations in δE as a function of delay time δt
- frequency changes with laser intensity

Interpretation:

order parameter amplitude oscillations



 $|2\Delta| \simeq 3 \text{ meV}$

How does a quantum system thermalize?

As $t \to \infty$, generic observables \hat{O} become time independent

True relaxation vs. decoherence:

- -- Decoherence: Only averaged observables become time-independent
 - non-interacting systems, integrable systems *only* show decoherence
- -- True relaxation: Generic local (unaveraged) observables become time-independent

 $\bar{O} = \lim_{t \to \infty} \langle \Psi(t) | \hat{O} | \Psi(t) \rangle$ time independent

 (i) systems coupled to bath, (ii) closed systems with certain interactions (infinite closed systems; otherwise recurrence)

Open questions:

- -- Which type of interactions lead to thermalization?
- -- Which observables \hat{O} to consider?
- -- How to describe thermalized sate? (is there a density matrix (ensemble) ρ such that: $\bar{O} = \text{Tr}[\hat{O} \rho]$?)

SC coupled to laser field and optical phonons

Goal: simulate *non-adiabatic* dynamics of superconductor coupled to (i) pump laser field and (ii) optical phonons

Microscopic model:



für Festkörperforschung

$$\begin{split} H &= \sum_{H=0}^{e_{1}} \sum_{\mathbf{k}s}^{+} c_{\mathbf{k}s} + \sum_{\mathbf{k}} \left(\left(\Delta_{\mathbf{k}} c_{\mathbf{k}\uparrow}^{+} c_{-\mathbf{k}\downarrow}^{+} + \mathbf{c}.\mathbf{c} \right) \right) \\ &- \frac{e}{r} - \frac{e\hbar}{m} \sum_{\mathbf{k}qs} (\mathbf{k} \cdot \mathbf{A}_{\mathbf{q}}) c_{\mathbf{k}+\frac{\mathbf{q}}{2}s}^{+} c_{\mathbf{k}-\frac{\mathbf{q}}{2}s} + \frac{e^{2}}{2m} \sum_{\mathbf{k}qs} (\mathbf{A}_{\mathbf{q}-\mathbf{k}} \cdot \mathbf{A}_{\mathbf{q}}) c_{\mathbf{k}s}^{+} c_{\mathbf{k}s} \\ &+ \sum_{\mathbf{q}j} + \sum_{\mathbf{q}j} \hbar \omega_{\mathbf{q}j} \left(b_{\mathbf{q}j}^{+} b_{\mathbf{q}j} + \frac{1}{2} \right) + \sum_{\substack{\mathbf{p}j\mathbf{k}s\\\mathbf{p}j\mathbf{k}s}} \left(g_{\mathbf{p}\mathbf{k}js} (b_{-\mathbf{p}j}^{+} + b_{\mathbf{p}j}) c_{\mathbf{k}+\mathbf{p},s}^{+} c_{\mathbf{k}s} + \mathbf{c}.\mathbf{c} \right) \right) + \mathbf{c.c.} \right) \end{split}$$

Goal: simulate *non-adiabatic* dynamics of superconductor coupled to optical phonons

- \implies considered hierarchies of time-scales: $\tau_p \ll \tau_{ph}, \tau_{\Delta} \ll \tau_{\epsilon}$



- time-dependent Ginzburg Landau theory, mu*-, and T*- models:
 - quasi-equilibrium is assumed at all times
 - time evolution of a single collective order parameter
 - only valid for: $\tau_{\Delta} \gg \tau_{\epsilon}$ (i.e., close to Tc, very dirty SCs)
 - Boltzmann kinetic equation:
 - requires adiabaticity on all time-scales
 - does not capture coherent evolution of quasi-particle distributions
 - only valid for: $\tau_{\rm p} \gg \tau_{\Delta}, \tau_{\rm ph}$

Phenomenological Rothwarf-Taylor models:

- rate equations for quasi-particle and phonon occupations
- only valid for: $\tau_{\rm p} \gg \tau_{\Delta}, \tau_{\rm ph}$

 \Rightarrow instead use density matrix formalism (expansion in $~\sim g_{
m ph}t/\hbar$)

$$\frac{d}{dt}\mathcal{O} = \frac{i}{\hbar} \left[H, \mathcal{O} \right]$$

Density matrix formalism: Equations of motion

Superconducting state: Bogoliubov transformation

$$\alpha_{\mathbf{k}}^{\dagger} = u_{\mathbf{k}}c_{\mathbf{k}\uparrow}^{\dagger} + v_{\mathbf{k}}^{*}c_{-\mathbf{k}\downarrow} \qquad \beta_{\mathbf{k}}^{\dagger} = u_{\mathbf{k}}c_{-\mathbf{k}\downarrow}^{\dagger} - v_{\mathbf{k}}^{*}c_{\mathbf{k}\uparrow}$$
$$\Delta_{\mathbf{k}} = \sum_{\mathbf{k}} W_{\mathbf{k},\mathbf{k}'} \left[u_{\mathbf{k}'}v_{\mathbf{k}'} \left(1 - \left\langle \alpha_{\mathbf{k}'}^{\dagger}\alpha_{\mathbf{k}'} \right\rangle - \left\langle \beta_{\mathbf{k}'}^{\dagger}\beta_{\mathbf{k}'} \right\rangle \right) + u_{\mathbf{k}'}^{2} \left\langle \beta_{\mathbf{k}'}\alpha_{\mathbf{k}'} \right\rangle - v_{\mathbf{k}'}^{2} \left\langle \alpha_{\mathbf{k}'}^{\dagger}\beta_{\mathbf{k}'}^{\dagger} \right\rangle \right]$$

All quantities of interest can be expressed in terms of these dynamical variables

$$\left\langle \alpha_{\boldsymbol{k}}^{\dagger} \alpha_{\boldsymbol{k}'} \right\rangle (t), \quad \left\langle \beta_{\boldsymbol{k}}^{\dagger} \beta_{\boldsymbol{k}'} \right\rangle (t), \quad \left\langle \alpha_{\boldsymbol{k}}^{\dagger} \beta_{\boldsymbol{k}'}^{\dagger} \right\rangle (t), \quad \left\langle \alpha_{\boldsymbol{k}} \beta_{\boldsymbol{k}'} \right\rangle (t) \quad \left\langle b_{\mathbf{p}} \right\rangle (t), \quad \left\langle b_{\mathbf{p}}^{\dagger} \right\rangle (t), \quad \left\langle b_{\mathbf{p}}^{\dagger} \right\rangle (t)$$

Current density:

$$\boldsymbol{j}(\boldsymbol{q},\omega) \simeq \frac{e\hbar}{mV} \sum_{\boldsymbol{k}} \boldsymbol{k} \left[\left\langle \alpha_{\boldsymbol{k}}^{\dagger} \alpha_{\boldsymbol{k}+\boldsymbol{q}} \right\rangle - \left\langle \beta_{\boldsymbol{k}+\boldsymbol{q}}^{\dagger} \beta_{\boldsymbol{k}} \right\rangle + \boldsymbol{k} \cdot \boldsymbol{q} \frac{\hbar^2 \left| \Delta_1 \right|}{2m \left(E_{\boldsymbol{k}}^2 \right)} \left(\left\langle \alpha_{\boldsymbol{k}}^{\dagger} \beta_{\boldsymbol{k}+\boldsymbol{q}}^{\dagger} \right\rangle + \left\langle \alpha_{\boldsymbol{k}+\boldsymbol{q}} \beta_{\boldsymbol{k}} \right\rangle \right) \right]$$

• Lattice displacement:
$$U(\mathbf{r},t) = \sqrt{\frac{\hbar}{2M\omega_{\rm ph}V}\sum_{\mathbf{p}}D_{\mathbf{p}}(t)e^{+i\mathbf{p}\cdot\mathbf{r}}},$$

with the *coherent phonon amplitude*: $D_{\mathbf{p}}(t) = \langle b_{\mathbf{p}} \rangle + \langle b_{-\mathbf{p}}^{\dagger} \rangle$

Density-matrix theory:

$$\frac{d}{dt}\mathcal{O} = \frac{i}{\hbar} \left[H, \mathcal{O} \right]$$

 \Rightarrow yields equations of motions for the above expectation values

Density matrix formalism: Equations of motion

Density-matrix theory:

$$\frac{d}{dt}\mathcal{O} = \frac{i}{\hbar} \left[H, \mathcal{O} \right]$$

 \Rightarrow yields equations of motions for

 $\left\langle \alpha_{\boldsymbol{k}}^{\dagger} \alpha_{\boldsymbol{k}'} \right\rangle (t), \quad \left\langle \beta_{\boldsymbol{k}}^{\dagger} \beta_{\boldsymbol{k}'} \right\rangle (t), \quad \left\langle \alpha_{\boldsymbol{k}}^{\dagger} \beta_{\boldsymbol{k}'}^{\dagger} \right\rangle (t), \quad \left\langle \alpha_{\boldsymbol{k}} \beta_{\boldsymbol{k}'} \right\rangle (t) \quad \left\langle b_{\mathbf{p}} \right\rangle (t), \quad \left\langle b_{\mathbf{p}}^{\dagger} \right\rangle (t), \quad \left\langle b_{\mathbf{p}}^{\dagger} \right\rangle (t)$

For example

$$i\hbar\frac{d}{dt}\left\langle\alpha_{\mathbf{k}}^{\dagger}\beta_{\mathbf{k}+\mathbf{q}}^{\dagger}\right\rangle = -\left(R_{\mathbf{k}}+R_{\mathbf{k}+\mathbf{q}}\right)\left\langle a_{\mathbf{k}}^{\dagger}\beta_{\mathbf{k}+\mathbf{q}}^{\dagger}\right\rangle + C_{\mathbf{k}+\mathbf{q}}^{\ast}\left\langle\alpha_{\mathbf{k}}^{\dagger}\alpha_{\mathbf{k}+\mathbf{q}}\right\rangle + C_{\mathbf{k}}^{\ast}\left(\left\langle\beta_{\mathbf{k}+\mathbf{q}}^{\dagger}\beta_{\mathbf{k}}\right\rangle - \delta_{\mathbf{q},0}\right)\right)$$
$$+\frac{e\hbar}{2m}\sum_{\mathbf{q}'=\pm\mathbf{q}_{0}}2\mathbf{k}\cdot\mathbf{A}_{\mathbf{q}'}(t)\left\{-L_{\mathbf{k},\mathbf{q}'}^{\dagger}\left\langle\alpha_{\mathbf{k}+\mathbf{q}'}^{\dagger}\beta_{\mathbf{k}+\mathbf{q}}^{\dagger}\right\rangle + L_{\mathbf{k}+\mathbf{q},-\mathbf{q}'}^{\dagger}\left\langle\alpha_{\mathbf{k}}^{\dagger}\beta_{\mathbf{k}+\mathbf{q}-\mathbf{q}'}^{\dagger}\right\rangle$$
$$-M_{\mathbf{k}+\mathbf{q},-\mathbf{q}'}^{-}\left\langle\alpha_{\mathbf{k}}^{\dagger}\alpha_{\mathbf{k}+\mathbf{q}-\mathbf{q}'}\right\rangle + M_{\mathbf{k},\mathbf{q}'}^{-}\left(\left\langle\beta_{\mathbf{k}+\mathbf{q}}^{\dagger}\beta_{\mathbf{k}+\mathbf{q}'}\right\rangle - \delta_{\mathbf{q},\mathbf{q}'}\right)\right\} + \cdots$$

Up to order $\left(m{A}_{m{q}}
ight)^n$, laser field only couples $\, (m{k},\,m{k}')$ to $\, (m{k},\,m{k}'+nm{q})$

⇒ leads to an effectively one-dimensional system of equations ⇒ interaction with laser field can be computed essentially exactly. [Papenkort, Kuhn, Axt, PRB 08]

Response of superconductor (w/o phonons)

- Two regimes:
- Adiabatic behavior for $~~ au_\Delta \ll au_{
 m p}$
- Non-adiabatic behavior for $~~ au_{
 m p} \ll au_{\Delta}$
- > Algebraically damped order parameter oscillations after short pump pulse ($\tau_{\rm p} \ll \tau_{\Delta}$):

$$|\Delta| = |\Delta_{\infty}| + \Gamma \frac{\cos(\frac{2|\Delta_{\infty}|}{\hbar}t + \Phi)}{\sqrt{t}}$$

("Higgs" amplitude mode)

[Volkov, Kogan, JETP 74] [Yuzbashyan, Altshuler PRL 06]







Algebraically damped oscillations due to decoherence

Order parameter oscillations

 $\tau_{\rm p} = 400 \text{ fs}$

 $\tau_{\rm p} = 10000 \, {\rm fs}$

20

10

t (ps)

()

30

40

 $-- \tau_{\rm p} = 1500 \, {\rm fs}$

1.6

1.4

1.2

1.0

0.8

-10

 $|\Delta(t)|$ (meV

Algebraically damped oscillations in averaged quantities due to decoherence

$$\Delta = W_0 \sum_{k \in \mathcal{W}} \langle c_{-k\downarrow} c_{+k\uparrow} \rangle$$
$$j_{q_0} = \frac{-e\hbar}{2mV} \sum_{k,\sigma} (2k+q_0) \langle c_{k,\sigma}^{\dagger} c_{k+q_0,\sigma} \rangle$$

But no true relaxation!

Quasiparticle occupations



Gap oscillations: Comparison with experiment

Qualitative agreement between theory and experiment

Numerical simulations:





Matsunaga, Shimano, et al. PRL 111, 057002 (2013)

THz-pump THz-probe spectroscopy NbN:

Pump-probe conductivity shows signatures of non-adiabatic dynamics

 $\sigma(\delta t, \omega) = j(\omega) / [i\omega A(\omega)]$

 \Rightarrow oscillations in pump-probe response as a function of delay time δt



Superconductor coupled to optical phonons

Linear coupling to optical phonons:

$$H_{\text{el-ph}} = g_{\text{ph}} \sum_{\mathbf{p}, \mathbf{k}, \sigma} \left[(b_{-\mathbf{p}}^{\dagger} + b_{\mathbf{p}}) c_{\mathbf{k}+\mathbf{p}, \sigma}^{\dagger} c_{\mathbf{k}, \sigma} + \text{c.c.} \right]$$

- perform expansion in $g_{\rm ph}$ using $\frac{d}{dt}\mathcal{O} = \frac{i}{\hbar}\left[H,\mathcal{O}\right]$

Phonon e

 \Rightarrow infinite hierarchy of equations of motion

Study generation of coherent phonons:

- Break hierarchy at first order: $\langle \alpha^{\dagger}_{\mathbf{k}} \beta^{\dagger}_{\mathbf{k}'} b_{\mathbf{p}} \rangle \simeq \langle \alpha^{\dagger}_{\mathbf{k}} \beta^{\dagger}_{\mathbf{k}'} \rangle \langle b_{\mathbf{p}} \rangle$

- Non-vanishing
$$\langle b_{\mathbf{p}} \rangle$$
 leads to finite lattice displacement: $U(\mathbf{r},t) = \sqrt{\frac{\hbar}{2M\omega_{\mathrm{ph}}V}} \sum_{\mathbf{p}} D_{\mathbf{p}}(t)e^{+i\mathbf{p}\cdot\mathbf{r}},$

with
$$D_{\mathbf{p}} = (b^{\dagger}_{-\mathbf{p}} + b_{\mathbf{p}})$$

- Equation of motion for coherent phonon amplitude: with forcing term:

 $\mathcal{F}_{\mathbf{p}}(t) = -\frac{2\omega_{\mathrm{ph}}}{\hbar}g_{\mathrm{ph}}\sum_{\mathbf{k}} \left[M_{\mathbf{k},\mathbf{p}}^{+} \left(\langle \alpha_{\mathbf{k}+\mathbf{p}}\beta_{\mathbf{k}}\rangle - \langle \alpha_{\mathbf{k}}^{\dagger}\beta_{\mathbf{k}+\mathbf{p}}^{\dagger}\rangle \right) + L_{\mathbf{k},\mathbf{p}}^{-} \left(\langle \alpha_{\mathbf{k}}^{\dagger}\alpha_{\mathbf{k}+\mathbf{p}}\rangle + \langle \beta_{\mathbf{k}+\mathbf{p}}^{\dagger}\beta_{\mathbf{k}}\rangle \right) \right],$

forced harmonic oscillator

$$\left[\frac{d^2}{dt^2} + \omega_{\rm ph}^2\right] D_{\bf p}(t) = \mathcal{F}_{\bf p}(t),$$

Equation of motion for coherent phonon amplitude

- Equation of motion for coherent phonon amplitude:

forced harmonic oscillator

$$\left[\frac{d^2}{dt^2} + \omega_{\rm ph}^2\right] D_{\mathbf{p}}(t) = \mathcal{F}_{\mathbf{p}}(t), \qquad \mathcal{F}_{\mathbf{p}}(t) = -\frac{2\omega_{\rm ph}}{\hbar} g_{\rm ph} \sum_k \left[M_{k,p}^+ \left(\langle \alpha_{k+p}\beta_k \rangle - \langle \alpha_k^\dagger \beta_{k+p}^\dagger \rangle\right) + \cdots\right]$$

Hierarchy of time scales:

 $\tau_{\rm p} \ll \tau_{\Delta} \ll \tau_{\rm ph}$

 $^{\prime}$ step-function

 $\Rightarrow \text{ Forcing term can be approximated by } \mathcal{F}_{\mathbf{p}}(t) \simeq A_{\mathbf{p}} \Theta(t)$ $\Rightarrow D_{\mathbf{p}}(t) \sim \frac{A_{\mathbf{p}}}{[1 - \cos(t) - t)]}$

$$\Rightarrow D_{\mathbf{p}}(t) \simeq \frac{m_{\mathbf{p}}}{\omega_{\mathrm{ph}}^2} [1 - \cos(\omega_{\mathrm{ph}}t)]$$

- abrupt change in quasiparticle states leads to jump in equilibrium position of lattice
- cosine oscillations in lattice displacement
- extrema at integer and half-integer values of $au_{
 m ph}$



Generation of coherent phonons: Numerical results

Hierarchy of time scales:

$$\tau_{\rm p} \ll \tau_{\Delta} \ll \tau_{\rm ph} \quad \Rightarrow \quad D_{\rm p}(t) \simeq \frac{A_{\rm p}}{\omega_{\rm ph}^2} [1 - \cos(\omega_{\rm ph} t)]$$

Numerical results for displacive excitation of coherent phonons:



- cosine oscillations in lattice displacement $U(R_0, t)$

- extrema at integer and half-integer values of τ_{ph}

APS, Manske, Avella, PRB 84, 214513 (2011)

Generation of coherent phonons: Quantum beats

Hierarchy of time scales: $au_{
m p} \ll au_{\Delta} \sim au_{
m ph}$

 \Rightarrow forcing term can be approximated by $\mathcal{F}_{\mathbf{p}}(t) \simeq \Theta(t) [A_{\mathbf{p}} + B_{\mathbf{p}} \cos(2\Delta_{\infty} t/\hbar)/\sqrt{t}],$

$$\Rightarrow \text{ quantum beating when } \omega_{\rm d} = |\omega_{\Delta_{\infty}} - \omega_{\rm ph}| \ll \omega_{\rm ph}$$
$$\Rightarrow D_{\mathbf{p}}(t) \simeq \frac{B_{\mathbf{p}}}{\omega_{\rm ph}} \sqrt{\frac{\pi}{2\omega_{\rm d}}} [\cos(t\omega_{\rm ph})S_2(t\omega_{\rm d}) + \sin(t\omega_{\rm ph})C_2(t\omega_{\rm d})],$$



Generation of coherent phonons: Pump-probe conductivity

Hierarchy of time scales: $au_{
m p} \ll au_{\Delta} \sim au_{
m ph}$

Signatures of non-adiabatic dynamics in pump-probe conductivity



beating phenomenon in pump-probe conductivity $\sigma(\delta t, \omega = \omega_{\rm ph})$ as a function of delay time δt

arXiv:1309.7318 (2013)

Generation of coherent phonons: Resonance

Hierarchy of time scales:

 $\tau_{\rm p} \ll \tau_{\Delta} = \tau_{\rm ph}$

 \Rightarrow resonant generation of coherent phonons for $\omega_{\rm ph} = \omega_{\Delta_{\infty}} (= 2\Delta_{\infty}/\hbar)$

$$\Rightarrow D_{\mathbf{p}}(t) \simeq \frac{B_{\mathbf{p}}}{\omega_{\mathrm{ph}}} \sqrt{t} \sin(\omega_{\mathrm{ph}}t) + \cdots$$



arXiv:1309.7318 (2013)

Conclusions & Outlook

- Microscopic simulation of ultrafast dynamics in superconductors
- > Non-adiabatic regime $au_{
 m p} \ll au_{\Delta}, au_{
 m ph}$:
- order parameter oscillations
 - \Rightarrow qualitative agreement with experiment
- generation of coherent phonos
- for $\ \hbar \omega_{\rm ph} = 2 \Delta_\infty$: resonant enhancement of coherent phonons
- Pump-probe conductivity:
- oscillations in $\sigma(\delta t, \omega)$ as a function of delay time with frequencies $\omega_{\Delta_{\infty}} = 2\Delta_{\infty}/\hbar$ and $\omega_{\rm ph}$
- strong enhancement of oscillation amplitude when frequencies are in resonance: $\omega_{\Delta_{\infty}} = \omega_{\rm ph}$

Outlook:

- consider higher order in correlation expansion: incoherent phonons, feedback on SC condensate



PRB 84, 214513 (2011); EPL 101, 17002 (2012); arXiv:1309.7318 (2013)